

# Quantum Stabilization of the 1/3-Magnetization Plateau in $\text{Cs}_2\text{CuBr}_4$

Jason Alicea,<sup>1</sup> Andrey V. Chubukov,<sup>2</sup> and Oleg A. Starykh<sup>3</sup>

<sup>1</sup>*Department of Physics, California Institute of Technology, Pasadena, California 91125, USA*

<sup>2</sup>*Department of Physics, University of Wisconsin, Madison, Wisconsin 53706, USA*

<sup>3</sup>*Department of Physics, University of Utah, Salt Lake City, Utah 84112, USA*

(Received 24 September 2008; revised manuscript received 5 November 2008; published 30 March 2009)

We consider the phase diagram of a spatially anisotropic 2D triangular antiferromagnet in a magnetic field. Classically, the ground state is umbrellalike for all fields, but we show that the quantum phase diagram is much richer and contains a 1/3-magnetization plateau, two commensurate planar states, two incommensurate chiral umbrella phases, and, possibly, a spin density wave state separating the two chiral phases. Our analysis sheds light on several recent experimental findings for  $\text{Cs}_2\text{CuBr}_4$ .

DOI: 10.1103/PhysRevLett.102.137201

PACS numbers: 75.10.Jm, 75.30.Ds, 75.50.Ee

**Introduction.**—A defining characteristic of frustrated quantum magnets is the appearance of numerous competing orders. This competition dramatically enhances quantum fluctuations, generating highly nonclassical behavior as exemplified by, e.g.,  $\text{Cs}_2\text{CuCl}_4$  and  $\text{Cs}_2\text{CuBr}_4$ . These materials comprise quasi-2D spin-1/2 triangular antiferromagnets with spatially anisotropic exchange [see Fig. 1(a)] and weak Dzyaloshinskii-Moriya (DM) coupling. Absent the latter, both systems classically should realize a zero-field coplanar spiral, which evolves into noncoplanar “umbrella” states in the field as in Fig. 1(b) with smoothly increasing magnetization up to saturation [1]. Experiments, however, reveal decidedly different behavior. In fields directed along the triangular layers,  $\text{Cs}_2\text{CuCl}_4$  realizes commensurate order in a wide field range [2,3], and  $\text{Cs}_2\text{CuBr}_4$  exhibits collinear “up-up-down” (UUD) order shown in Fig. 1(c) over a finite field interval, yielding a 1/3-magnetization plateau [4–7]. At its boundaries, the UUD phase undergoes first-order transitions into *planar* states [6–9]. Further experiments [6,10] on  $\text{Cs}_2\text{CuBr}_4$  suggest the presence of a narrow 2/3 plateau and other intervening collinear phases as well.

While the UUD phase is well established for the isotropic triangular antiferromagnet, much less is known about the plateau’s stability and its proximate quantum phases in the anisotropic case. The challenge here is illuminated by first observing that when  $J = J'$ , the UUD state appears due to an “accidental” classical degeneracy between umbrella and planar states of Figs. 1(b) and 1(c), which quantum fluctuations lift in favor of the latter [11]. When  $J \neq J'$ , this degeneracy is lifted already at the classical level but in favor of *umbrella* states for all fields. Planar order can then emerge only if quantum effects overshadow those of spatial anisotropy. The standard spin-wave expansion is, however, not suitable for studying this competition since the planar phases cease to be classical ground states. To address the quantum phase diagram for the anisotropic system, particularly near 1/3 magnetization, we employ a modified approach which is controlled

by the smallness of  $1/S$  and spatial anisotropy and yields nonanalytic results in both parameters.

Figure 1(d) summarizes our results. Since classical degeneracy lifting is  $\propto (J - J')^2/J$  while quantum corrections are  $\propto J/S$ , the physics is conveniently described by the parameter  $\delta = (40/3)S(J - J')^2/J^2$ . For  $\delta < 1$ , the UUD phase’s stability is, counterintuitively, unaffected by anisotropy. The spin order adjacent to the plateau remains coplanar and *commensurate*, with incommensurate phases appearing only at small and high fields. For  $1 < \delta < 4$ , the UUD phase persists but at the boundaries becomes unstable toward noncoplanar, incommensurate “distorted umbrella” phases (this happens for  $\delta > 1$  at the lower boundary and for  $\delta > 3$  at the upper boundary). These two phases emerge as finite- $k$  instabilities of different spin-wave branches of the UUD phase, and both have a nonzero Ising chirality order parameter  $K_{ABC} = \hat{z} \cdot (\mathbf{S}_A \times \mathbf{S}_B + \mathbf{S}_B \times \mathbf{S}_C + \mathbf{S}_C \times \mathbf{S}_A)$  for each plaquette. While the

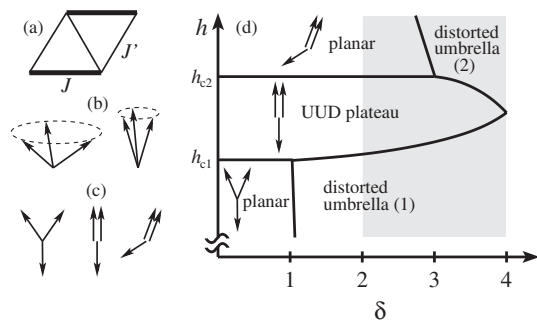


FIG. 1. (a) Anisotropic triangular lattice with exchanges  $J$  and  $J'$ . (b) Umbrella and (c) planar phases comprise competing classical ground states of the isotropic nearest-neighbor Heisenberg model. (d) Proposed quantum phase diagram for the *anisotropic* model near 1/3 magnetization (full field range not shown). The horizontal axis is  $\delta = (40/3)S(J - J')^2/J^2$ . Planar states shown are *commensurate*, though incommensurate states are predicted at small and large fields. The shaded area is where the UUD and adjacent phases are metastable, the energy being minimized by umbrella states in (b).

UUD phase ceases to exist for  $\delta > 4$ , the distorted umbrellas must remain separated in this regime since their chiralities are uncorrelated. We show that at  $\delta = 4$  the system has an extended symmetry, which allows both one-magnon and two-magnon excitations to develop. Two-magnon condensation leads to an intermediate *collinear* spin density wave (SDW) phase found previously in the opposite limit of small  $J'/J$  [12]. This agreement and the fact that experiments on the more anisotropic  $\text{Cs}_2\text{CuCl}_4$  [2,13] support collinear SDW order for 1–3 T fields make this SDW a plausible intermediate phase between the low- and high-field distorted umbrellas for  $\delta > 4$ .

As a further complication, in the shaded region of Fig. 1(d) with  $\delta > 2$  the UUD state has higher energy than the classical, undistorted umbrella; i.e., for  $2 < \delta < 4$ , the UUD state and neighboring phases are *metastable*. These may still be probed in pulsed field experiments [14].

*Model and UUD state in the anisotropic system.*—We consider a simple Heisenberg model with

$$H = \sum_{\langle \mathbf{r}\mathbf{r}' \rangle} J_{\mathbf{r}\mathbf{r}'} \mathbf{S}_{\mathbf{r}} \cdot \mathbf{S}_{\mathbf{r}'} - hS \sum_{\mathbf{r}} S_{\mathbf{r}}^z, \quad (1)$$

where  $\mathbf{S}_{\mathbf{r}}$  are spin- $S$  operators, the exchanges  $J_{\mathbf{r}\mathbf{r}'}$  are as shown in Fig. 1(a), and  $h$  is the (scaled) magnetic field. The saturation field is  $h_{\text{sat}} = (2J + J')^2/J$ . To treat quantum and anisotropy effects on equal footing, we organize our analysis by assuming small  $(J - J')/J$  and  $1/S$ . Interlayer exchange and DM coupling will be present in the materials but are weak [15] and will be neglected (though we restore DM below). Use of the large- $S$  expansion for  $S = 1/2$  systems is an approximation, but it generally works well for magnetically ordered states which we consider ( $\text{Cs}_2\text{CuBr}_4$  exhibits magnetic order at all fields).

With  $J = J'$ , the commensurate (three-sublattice) umbrella and planar states of Figs. 1(b) and 1(c) are classically degenerate. Quantum fluctuations favor planarity, and spin rearrangement in a field occurs as in Fig. 1(c). This process includes an intermediate UUD phase, which is classically stable only at  $h_{\text{sat}}/3$ . Quantum fluctuations, which generally favor collinear states [16,17], extend its stability to a finite field interval  $h_{c1}^0 \leq h \leq h_{c2}^0$  [11], resulting in a  $1/3$ -magnetization plateau. To leading order in  $1/S$

$$h_{c1}^0 = 3J - \frac{0.50J}{2S}, \quad h_{c2}^0 = 3J + \frac{1.3J}{2S}, \quad (2)$$

which for  $S = 1/2$  yields a plateau in a range  $\Delta h^0 = h_{c2}^0 - h_{c1}^0 = 1.8J/(2S)$ , in good agreement with exact diagonalization [18]. Inside this range, there are two low-energy spin-wave modes with gaps  $\propto |h_{c1,2}^0 - h|$  at  $\mathbf{k} = 0$ .

When  $J \neq J'$ , the umbrella state becomes incommensurate and classically has lower energy than the planar phase for all fields. To study the stability of the classically unfavorable UUD state, we explore a modified large- $S$  approach to Eq. (1). First, we use a three-sublattice representation, where spins point up on sublattices  $A$  and  $B$  and down on sublattice  $C$ , and introduce Holstein-Primakoff bosons  $a$ ,  $b$ , and  $c$ , respectively. The linear spin-wave

Hamiltonian so obtained is problematic due to the classical instability of harmonic spin waves at  $\delta \neq 0$ . However, the *interacting* spin-wave Hamiltonian must support a stable UUD plateau over a finite  $\delta$  range, as exact diagonalization finds [19]. Therefore, we extend the linear spin-wave Hamiltonian of the UUD state to include the leading  $1/S$  self-energy corrections obtained by decoupling 4-boson interactions using correlations from the *isotropic* system:

$$H_{\text{uud}} = S \sum_{\mathbf{k}} \{ [\gamma_{1,\mathbf{k}} a_{\mathbf{k}}^\dagger b_{\mathbf{k}} + \gamma_{2,\mathbf{k}} (b_{\mathbf{k}}^\dagger c_{-\mathbf{k}}^\dagger + c_{\mathbf{k}}^\dagger a_{-\mathbf{k}}^\dagger) + \text{H.c.}] \\ + (h + \Sigma_1)(a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + b_{\mathbf{k}}^\dagger b_{\mathbf{k}}) + (2h_0 + \Sigma_2 - h)c_{\mathbf{k}}^\dagger c_{\mathbf{k}} \}. \quad (3)$$

Here the summation extends over the magnetic Brillouin zone, and we have defined  $h_0 = J + 2J'$  and

$$\gamma_{j,\mathbf{k}} = \Sigma'_{j,\mathbf{k}} + J e^{ik_x} + 2J' \cos(\sqrt{3}k_y/2) e^{-ik_x/2}. \quad (4)$$

The self-energy components are  $\Sigma_1 = 0.14J/S$ ,  $\Sigma_2 = 0.67J/S$ ,  $\Sigma'_{1,0} = -0.11J/S$ , and  $\Sigma'_{2,0} = 0.18J/S$ . Low-energy excitations near  $\mathbf{k} = 0$  encode the important physics, and in this region analysis of Eq. (3) simplifies considerably. Here we have  $\gamma_{j,\mathbf{k}} \approx \tilde{\gamma}_{j,\mathbf{k}} + i\Gamma_{\mathbf{k}}$ , with

$$\tilde{\gamma}_{j,\mathbf{k}} = h_0 + \Sigma'_{j,0} - \frac{3}{4}Jk^2, \quad \Gamma_{\mathbf{k}} = (J - J')k_x. \quad (5)$$

Diagonalizing Eq. (3) with  $\Gamma_{\mathbf{k}} = 0$  yields

$$H'_{\text{uud}} = S \sum_{\mathbf{k}} [\omega_p p_{\mathbf{k}}^\dagger p_{\mathbf{k}} + \omega_v v_{\mathbf{k}}^\dagger v_{\mathbf{k}} + \omega_u u_{\mathbf{k}}^\dagger u_{\mathbf{k}}]. \quad (6)$$

The  $u$  bosons describe precession of the total magnetization and have a large gap  $\omega_u(\mathbf{0}) \sim h$ , while the  $p$  and  $v$  bosons are the low-energy modes of interest. For small  $\mathbf{k}$  we obtain

$$\omega_p(\mathbf{k}) = [h - h_{c1}^0 + 2(J - J')] + \frac{3}{4}Jk^2, \quad (7) \\ \omega_v(\mathbf{k}) \approx [h_{c2}^0 - 2(J - J') - h] + \frac{9}{4}Jk^2.$$

We now neglect the high-energy  $u$  mode. Terms involving  $\Gamma_{\mathbf{k}}$ , which we denote by  $H''_{\text{uud}}$ , then take a simple form:

$$H''_{\text{uud}} = iS \sum_{\mathbf{k}} \Gamma_{\mathbf{k}} (\sqrt{2} \cosh \theta_{\mathbf{k}} - \sinh \theta_{\mathbf{k}}) (p_{\mathbf{k}} v_{-\mathbf{k}} - \text{H.c.}), \quad (8)$$

where  $\theta_{\mathbf{k}}$  follows from  $\tanh(2\theta_{\mathbf{k}}) = -2\sqrt{2}\tilde{\gamma}_{2,\mathbf{k}}/(2h_0 + \Sigma_1 + \Sigma_2 + \tilde{\gamma}_{1,\mathbf{k}})$ . Diagonalizing  $H'_{\text{uud}} + H''_{\text{uud}}$  in the low-energy spin-wave sector, we obtain

$$\tilde{H}_{\text{uud}} = S \sum_{\mathbf{k}} [\omega_1 d_{1,\mathbf{k}}^\dagger d_{1,\mathbf{k}} + \omega_2 d_{2,\mathbf{k}}^\dagger d_{2,\mathbf{k}}]. \quad (9)$$

The leading small  $\mathbf{k}$  energies are

$$\omega_{1,2}(\mathbf{k}) = \pm \left( h - h_0 - \frac{1}{5S}J - \frac{3}{4}Jk^2 \right) + \frac{3JZ_{\mathbf{k}}}{20S}, \quad (10)$$

with  $Z_{\mathbf{k}} = \sqrt{9 + 10S(6k^2 - 3\delta k_x^2 + 10S\mathbf{k}^4)}$ . The UUD phase is stable for  $h_{c1} < h < h_{c2}$ , where

$$h_{c1,2} = h_{c1,2}^0 + 2(J' - J) \mp \frac{3J}{4} \min \left( \mp k_x^2 + \frac{Z_{\mathbf{k}} - 3}{5S} \right). \quad (11)$$

Equations (10) and (11), which are nonanalytic in  $1/S$  and  $J - J'$ , dictate the UUD state's local stability in the anisotropic system. One can verify by sending  $S \rightarrow \infty$  above that the UUD state is indeed unstable for arbitrary anisotropy in the classical limit, due to a finite- $k_x$  instability. For  $\delta < 1$ , both  $\omega_{1,2}$  are minimized at  $\mathbf{k} = 0$ , so it follows from Eq. (11) that the plateau width  $\Delta h$  is unchanged from the isotropic system.

Surprisingly, in the quantum system a finite amount of anisotropy is required to begin destabilizing the plateau. For  $\delta > 1$ , the minimum of  $\omega_1$  shifts to  $\mathbf{k}_{1\pm} = (\pm k_1, 0)$ , where  $k_1^2 = [3\delta - 6 + \sqrt{3\delta(4 - \delta)}]/(20S)$ ;  $h_{c1}$  then moves upward, reducing the plateau width. Similarly, for  $\delta > 3$ , the minimum of  $\omega_2$  shifts to  $\mathbf{k}_{2\pm} = (\pm k_2, 0)$ , with  $k_2^2 = [3\delta - 6 - \sqrt{3\delta(4 - \delta)}]/(20S)$ . At this point,  $h_{c2}$  moves to a smaller value, further reducing the UUD region. The plateau ceases to be locally stable at  $\delta = 4$ , when both spin waves become gapless at  $k_1^2 = k_2^2 = k_m^2 = 3/(10S)$ .

*Proximate phases.*—We now explore the phases that emerge at  $h_{c1}$  and  $h_{c2}$ , where the UUD spin waves Bose condense. Here one must determine the order parameters that become nonzero at the transition and what this implies for the spin components  $\langle S^{x,y} \rangle$ . This is straightforward for  $\delta < 1$ : The minima of  $\omega_j$  ( $j = 1, 2$ ) occur at  $\mathbf{k} = 0$ , and the relevant order parameters are simply  $\psi_j \propto \langle d_{j,0} \rangle$ . One can easily verify that condensation of  $\psi_j$  at  $h = h_{cj}$  leads to the *commensurate* coplanar spin configurations displayed in Fig. 1(c). This prediction is rather nontrivial and could be tested in exact diagonalization studies.

The situation is subtler at  $h_{c1}$  when  $\delta > 1$ . Here  $\omega_1(\mathbf{k})$  possesses two inequivalent minima at  $\mathbf{k}_{1\pm}$ , so there are two order parameters:  $\psi_{\pm} = \sqrt{3/NS} \langle d_{1,\mathbf{k}_{1\pm}} \rangle$  ( $N$  is the number of spins). An energy functional for  $\psi_{\pm}$  can be obtained by retaining quartic interactions between the  $d_1$  bosons in the spin-wave Hamiltonian [20]; we obtain

$$\frac{2E}{JNS^2} = r(|\psi_+|^2 + |\psi_-|^2) + (|\psi_+|^2 + |\psi_-|^2)^2 + u|\psi_+|^2|\psi_-|^2. \quad (12)$$

Here  $r \propto h - h_{c1}$  and  $u = 2\cosh^2 2\phi_{k_1}$ , where

$$\tanh(2\phi_{k_1}) = \frac{6(J - J')k_1}{\omega_1(k_1) + \omega_2(k_1)} = \sqrt{3\delta} \frac{\sqrt{10Sk_1}}{3 + 10Sk_1^2}. \quad (13)$$

Since  $u > 0$ , interactions favor  $\psi_+ \neq 0$ ,  $\psi_- = 0$ , or vice versa at the transition. Choosing the former, the spin configuration can be written  $\langle S_{A,B}^+ \rangle = S\psi_+(\pm \cosh\phi_{k_1} - i \sinh\phi_{k_1})e^{-ik_1x}$ ,  $\langle S_C^+ \rangle = 2iS\psi_+ \sinh\phi_{k_1}e^{-ik_1x}$ . This corresponds to noncoplanar, incommensurate order that can be described as a distorted umbrella. This state has a finite chirality, whose sign is determined by the condensate momentum via  $K_{ABC}^{(1)} = \pm 3S^2|\psi_{\pm}|^2 \sinh 2\phi_{k_1}$ .

Analogous physics arises at  $h_{c2}$  when  $\delta > 3$ :  $\omega_2(\mathbf{k})$  has minima at  $\mathbf{k}_{2\pm}$ , and the energy has the same form as in (12) but with  $\bar{\psi}_{\pm} = \sqrt{3/NS} \langle d_{2,\mathbf{k}_{2\pm}} \rangle$ . The spin configura-

tion above  $h_{c2}$  is a distorted umbrella with chirality  $K_{ABC}^{(2)} = \mp 3S^2|\bar{\psi}_{\pm}|^2 \sinh 2\phi_{k_2}$ . Taking  $\bar{\psi}_{\pm} \neq 0$ , we have  $\langle S_{A,B}^+ \rangle = S\bar{\psi}_{\pm}(\mp \sinh\phi_{k_1} - i \cosh\phi_{k_1})e^{ik_1x}$ ,  $\langle S_C^+ \rangle = 2iS\bar{\psi}_{\pm} \cosh\phi_{k_1}e^{ik_1x}$ .

At  $\delta = 4$ , the UUD plateau shrinks to a point at  $h_c = h_0 + 17J/(40S)$  and becomes unstable at larger  $\delta$ . How the two distorted umbrellas merge in this regime presents an interesting issue. Since these states arise upon condensation of *different* spin-wave modes at  $h_{c1,2}$ , their chiralities are uncorrelated. The two phases then cannot gradually transform into each other and must be separated either by a first-order transition or by an intermediate phase with no chirality.

To gain insight here, we study the instabilities at  $\delta = 4$ ,  $h = h_c$ . At this point, both spin-wave modes become gapless at the same  $\pm k_m$ , and the coherence factors  $\sinh\phi_{k_m}$  and  $\cosh\phi_{k_m}$  diverge as  $1/\sqrt{4 - \delta}$ , so that  $\tanh 2\phi_{k_m} \rightarrow 1$ . We now have four order parameters:  $\psi_{\pm}$  and  $\bar{\psi}_{\pm}$ . The corresponding energy functional at  $\delta = 4$ ,  $h = h_c$  to fourth order in  $\psi_{\pm}$  and  $\bar{\psi}_{\pm}$  is

$$\frac{2E}{JNS^2} = (|\psi_+|^2 + |\psi_-|^2 - |\bar{\psi}_+|^2 - |\bar{\psi}_-|^2)^2 + 2|\psi_+|^2|\bar{\psi}_+|^2 + 2|\psi_-|^2|\bar{\psi}_-|^2, \quad (14)$$

subject to the constraint  $\psi_+ \psi_+^* - \bar{\psi}_- \bar{\psi}_-^* = i(\psi_+ \bar{\psi}_- + \psi_+^* \bar{\psi}_-^*)$  which eliminates infinitely large terms from the energy. Choosing just one order parameter nonzero, we obtain the same distorted umbrellas as before, with  $E \propto |\psi|^4$  and a finite chirality. However, we see from (14) that there is a better choice—taking  $|\psi_+| = |\bar{\psi}_-| \neq 0$ ,  $\psi_- = \bar{\psi}_+ = 0$ , or vice versa yields  $E = 0$ . Thus, unlike the situation elsewhere on the critical lines  $h_{c1,2}(\delta)$ , the magnitude of the condensate  $|\psi_+| = |\bar{\psi}_-|$  at  $\delta = 4$  is *not* constrained, implying an extended symmetry at this point. Specifically, the symmetry is  $U(1) \times U(1) \times P_1$ , where the two  $U(1)$ 's represent phases of the order parameters, while  $P_1$  reflects the unconstrained nature of their absolute values. Unusual structure at  $\delta = 4$ ,  $h = h_c$  also appears in the linear dispersion of  $\omega_{1,2}$  near the minima  $\pm k_m$ . Any other point on the UUD phase boundary has one gapless mode with quadratic dispersion near the spectrum's minima. The difference arises because at  $\delta < 4$  the condensate is zero at the critical point while at  $\delta = 4$  its value can be arbitrary (hence the  $P_1$  symmetry). Simultaneous breaking of the two  $U(1)$ 's drives nonplanar order whose precise structure depends on the relative phase of the two single-particle condensates. The extended symmetry uncovered by us offers a more intriguing possibility of two-particle condensation: This breaks  $P_1$  but preserves  $U(1)$  and leads to SDW order found previously in the limit  $J'/J \ll 1$  [12].

*Phase diagram.*—So far, we have analyzed the UUD phase's local stability without addressing whether it globally minimizes the energy. There are three regimes where one can easily compare the umbrella and planar energies. First is the high-field regime  $h \approx h_{\text{sat}}$ . There the umbrella



state, which at arbitrary  $h$  is described by

$$\mathbf{S}_{\mathbf{r}} = S\{\cos\theta[\cos(\mathbf{Q} \cdot \mathbf{r})\hat{\mathbf{x}} + \sin(\mathbf{Q} \cdot \mathbf{r})\hat{\mathbf{y}}] + \sin\theta\hat{\mathbf{z}}\}, \quad (15)$$

with  $\mathbf{Q} = 2\cos^{-1}(-J'/2J)$  and  $\sin\theta = h/h_{\text{sat}}$ , wins for all  $J' \neq J$  simply because quantum effects vanish at  $h_{\text{sat}}$ . We verified this explicitly by computing the analog of Eq. (12) at  $h_{\text{sat}}$  to show that indeed interactions drive the system into the umbrella state for arbitrary  $J' \neq J$ . The critical line which begins at  $\delta = 3$ ,  $h = h_{c2}$  thus ends up at  $\delta = 0$ ,  $h = h_{\text{sat}}$ .

The second regime occurs at small  $h \rightarrow 0$ . Here the lowest-energy planar configuration is *incommensurate*, with the same  $\mathbf{Q}$  as the umbrella state and

$$\mathbf{S}_{\mathbf{r}} = S[\cos(\mathbf{Q} \cdot \mathbf{r} + \varphi_{\mathbf{r}})\hat{\mathbf{z}} + \sin(\mathbf{Q} \cdot \mathbf{r} + \varphi_{\mathbf{r}})\hat{\mathbf{x}}], \quad (16)$$

where  $\varphi_{\mathbf{r}} = -(2h/u)\sin(\mathbf{Q} \cdot \mathbf{r}) + O(h^2)$  and  $u = h_{\text{sat}}[1 + (J - J')^2/J^2]$ . At small  $h$ , the energy difference between the umbrella and planar states of Eqs. (15) and (16) is  $\Delta E_{h \rightarrow 0} \equiv (E_{\text{umb}} - E_{\text{pl}})/NS^2 = -(1/2)h^2\Delta\chi$ , where  $\Delta\chi = \chi_{\text{umb}} - \chi_{\text{pl}}$  is the difference of susceptibilities. In the classical limit, we find  $\chi_{\text{umb}} = 1/h_{\text{sat}}$ ,  $\chi_{\text{pl}} = 1/u$ , so that  $\Delta\chi = (J - J')^2/(9J^3)$  and the umbrella state has lower energy. The competition comes from quantum fluctuations:  $1/S$  corrections to  $\chi_{\text{umb}}$  and  $\chi_{\text{pl}}$  are different already for  $J = J'$  and such that  $\Delta\chi_{\text{qu}} \approx -0.16/(18JS)$  [11]. Adding the two contributions, we find that  $\Delta E_{h \rightarrow 0} = [0.008h^2/(2JS)](1.1 - \delta)$ ; i.e., the incommensurate planar state has lower energy for  $\delta < 1.1$ . This implies that the commensurate planar state that we found immediately below  $h_{c1}$  undergoes a transition into an incommensurate planar state at  $h < h_{c1}$ . Thus, the line separating planar and distorted umbrella states at low fields departs at  $\delta = 1$ ,  $h = h_{c1}$  and ends up at  $\delta = 1.1$ ,  $h = 0$ .

Finally, at  $h_{\text{sat}}/3$  the energy difference between the umbrella and UUD phase is  $\Delta E_{1/3} = [0.067J/(2S)](2.0 - \delta)$ , where the first and second terms, respectively, are the classical and quantum contributions [11]. Consequently, the UUD phase and the neighboring distorted umbrella phases remain global minima only up to  $\delta = 2.0$ . For  $\delta > 2$ , the UUD state is metastable and observable only via a transient magnetization plateau, similar to the situation in a kagome system [14].

*Discussion.*—The resulting phase diagram near  $1/3$  magnetization is shown in Fig. 1(d). The shaded region denotes the regime where the classical umbrella minimizes the energy globally. This phase diagram is in agreement with data for  $\text{Cs}_2\text{CuBr}_4$ , where  $J'/J = 0.7$  implies that  $\delta = 0.6$  if we extrapolate to  $S = 1/2$ . For this  $\delta$ , the UUD state is present, and the nearby phases are planar, in agreement with NMR [8,9] and neutron [6] experiments. These experiments also observe that both transitions out of the UUD state are first order. Our calculations predict continuous transitions as a consequence of the U(1) spin

symmetry exhibited by the Hamiltonian (1). However, when this U(1) symmetry is broken explicitly by spin-orbit coupling, cubic terms in the energy functionals describing the transition [such as (12)] are permissible, which generically render the transition first order. In particular, DM coupling of the form present in  $\text{Cs}_2\text{CuBr}_4$  breaks this symmetry (and is allowed by momentum conservation for  $\delta < 1$ ) when the field is directed along the triangular layers. In addition, a direct first-order transition from the UUD phase into the incommensurate planar phase is also a possibility, which should be investigated by numerical calculations similar to those in Ref. [21]. For  $\text{Cs}_2\text{CuCl}_4$ , the anisotropy is much higher ( $\delta \approx 2.9$ ), and the system very likely lies outside of the applicability region of our analysis and should be approached from a 1D perspective [12]. Still, even within our framework,  $\delta > 2$  implies no UUD phase, and no plateau is seen in  $\text{Cs}_2\text{CuCl}_4$ .

We acknowledge illuminating discussions with O. Motrunich, M. Takigawa, and Y. Takano, who we also thank for sharing experimental data. This work was supported by the Lee A. DuBridge Foundation (J. A.) and by NSF-DMR 0604406 (A. V. C.).

- 
- [1] M. Y. Veillette, J. T. Chalker, and R. Coldea, Phys. Rev. B **71**, 214426 (2005).
  - [2] Y. Tokiwa *et al.*, Phys. Rev. B **73**, 134414 (2006).
  - [3] M. Y. Veillette and J. T. Chalker, Phys. Rev. B **74**, 052402 (2006).
  - [4] T. Ono *et al.*, Phys. Rev. B **67**, 104431 (2003).
  - [5] T. Ono *et al.*, J. Phys. Condens. Matter **16**, S773 (2004).
  - [6] T. Ono *et al.*, Prog. Theor. Phys. Suppl. **159**, 217 (2005).
  - [7] H. Tsuji *et al.*, Phys. Rev. B **76**, 060406(R) (2007).
  - [8] Y. Fujii *et al.*, Physica (Amsterdam) **346B–347B**, 45 (2004).
  - [9] Y. Fujii *et al.*, J. Phys. Condens. Matter **19**, 145237 (2007).
  - [10] N. A. Fortune, S. T. Hannahs, Y. Yoshida, T. E. Sherline, T. Ono, H. Tanaka, and Y. Takano, arXiv:0812.2077.
  - [11] A. V. Chubukov and D. I. Golosov, J. Phys. Condens. Matter **3**, 69 (1991); the lifting of the accidental degeneracy by classical, thermal fluctuations has been analyzed by H. Kawamura and S. Miyashita, J. Phys. Soc. Jpn. **54**, 4530 (1985).
  - [12] O. A. Starykh and L. Balents, Phys. Rev. Lett. **98**, 077205 (2007).
  - [13] M. Takigawa (private communication).
  - [14] Y. Narumi *et al.*, Europhys. Lett. **65**, 705 (2004).
  - [15] R. Coldea *et al.*, Phys. Rev. Lett. **88**, 137203 (2002).
  - [16] E. F. Shender, Sov. Phys. JETP **56**, 178 (1982).
  - [17] C. L. Henley, Phys. Rev. Lett. **62**, 2056 (1989).
  - [18] A. Honecker, J. Schulenburg, and J. Richter, J. Phys. Condens. Matter **16**, S749 (2004).
  - [19] S. Miyahara, K. Ogino, and N. Furukawa, Physica (Amsterdam) **378B–380B**, 587 (2006).
  - [20] T. Nikuni and S. Shiba, J. Phys. Soc. Jpn. **64**, 3471 (1995).
  - [21] S. Yoshikawa *et al.*, J. Phys. Soc. Jpn. **73**, 1798 (2004).